

Form PTO-1390 (Rev. 12-29-99)		US DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE	ATTORNEY'S DOCKET NO. H 3329 PCT/US
TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371			U.S. APPLICATION NO. (if known see 37 CFR 1.5) 09/744001
INTERNATIONAL APPLICATION NO. PCT/EP99/04836	INTERNATIONAL FILING DATE July 9, 1999	PRIORITY DATE CLAIMED July 18, 1998	
TITLE OF INVENTION METHOD FOR PREPARING ALCOXYLATED CARBOXYLIC ACID ESTERS			
APPLICANT(S) FOR DO/EO/US Ansgar BEHLER and Almud FOLGE			
Applicant herewith submits to the United States Designated/Elected Office (EO/DO/US) the following items and other information:			
<p>1. <input checked="" type="checkbox"/> This is a FIRST submission of items concerning a filing under 35 U.S.C. 371.</p> <p>2. <input type="checkbox"/> This is a SECOND or SUBSEQUENT submission of items concerning a filing under 35 U.S.C. 371.</p> <p>3. <input type="checkbox"/> This express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39 (1).</p> <p>4. <input checked="" type="checkbox"/> A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date.</p> <p>5. <input checked="" type="checkbox"/> A copy of the International Application as filed (35 U.S.C. 371(c)(2)).</p> <p>a. <input type="checkbox"/> is transmitted herewith (required only if not transmitted by the International Bureau).</p> <p>b. <input checked="" type="checkbox"/> has been transmitted by the International Bureau.</p> <p>c. <input type="checkbox"/> is not required, as the application was filed in the United States Receiving Office (RO/US).</p> <p>6. <input checked="" type="checkbox"/> A translation of the International Application into English (35 U.S.C. 371(c)(2)).</p> <p>7. <input checked="" type="checkbox"/> Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3)).</p> <p>a. <input type="checkbox"/> are transmitted herewith (required only if not transmitted by the International Bureau).</p> <p>b. <input type="checkbox"/> have been transmitted by the International Bureau.</p> <p>c. <input type="checkbox"/> have not been made; however, the time limit for making such amendments has NOT expired.</p> <p>d. <input checked="" type="checkbox"/> have not been made and will not be made.</p> <p>8. <input type="checkbox"/> A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).</p> <p>9. <input checked="" type="checkbox"/> An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)). (UNEXECUTED)</p> <p>10. <input type="checkbox"/> A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).</p>			
Items 11. to 16. below concern other document(s) or information included:			
<p>11. <input type="checkbox"/> An Information Disclosure Statement under 37 CFR 1.97 and 1.98.</p> <p>12. <input type="checkbox"/> An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.</p> <p>13. <input checked="" type="checkbox"/> A FIRST preliminary amendment</p> <p><input type="checkbox"/> A SECOND or SUBSEQUENT preliminary amendment.</p> <p>14. <input type="checkbox"/> A substitute specification.</p> <p>15. <input type="checkbox"/> A change of power of attorney and/or address letter.</p> <p>16. <input type="checkbox"/> Other items or information:</p>			
"Express Mail Post Office to Addressee" service Mailing Label Number			
<u>EL541613488US</u>			

U.S. Application No. (If known, see 37 CFR 1.55) 097744001	INTERNATIONAL APPLICATION NO. PCT/EP99/04836	ATTORNEY'S DOCKET NUMBER H 3329 PCT/US
17. The following fees are submitted: BASIC NATIONAL FEE (37 CFR 1.492(a)(1)-(5)): Neither International preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO and International Search Report not prepared by the EPO or JPO..... \$1,000.00 International preliminary examination fee (37 CFR 1.482) not paid to USPTO but International Search Report prepared by the EPO or JPO..... \$860.00 International preliminary examination fee (37 CFR 1.482) not paid to USPTO but international search fee (37 CFR 1.445(a)(2)) paid to USPTO \$710.00 International preliminary examination fee paid to USPTO (37 CFR 1.482) but all claims did not satisfy provisions of PCT Article 33(1)-(4) \$690.00 International preliminary examination fee paid to USPTO (37 CFR 1.482) and all claims satisfied provisions of PCT Article 33(1)-(4)..... \$100.00		CALCULATIONS PTO USE ONLY
ENTER APPROPRIATE BASIC FEE AMOUNT = \$ 860		
Surcharge of \$130.00 for furnishing the oath or declaration later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(e)).		\$ 0
CLAIMS	NUMBER FILED	NUMBER EXTRA
Total Claims	14 - 20 =	0
Independent Claims	1 - 3 =	0
Multiple dependent claims (s) (if applicable) 0 + \$270.00		\$ 0
TOTAL OF ABOVE CALCULATIONS = \$ 860		
Reduction of 1/2 for filing by small entity, if applicable. A Small Entity Statement must also be filed. (Note 37 CFR 1.9, 1.27, 1.28).		\$ 0
SUBTOTAL = \$ 860		
Processing fee of \$130.00 for furnishing the English translation later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(f)).		\$ 0
TOTAL NATIONAL FEE = \$ 860		
Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). \$40.00 per property		\$ 0
TOTAL FEES ENCLOSED = \$ 860		
		Amount to be refunded: \$-----
		charged: \$ 860.00
a. <input type="checkbox"/> A check in the amount of \$_____ to cover the above fees is enclosed. b. <input checked="" type="checkbox"/> Please charge my Deposit Account No. <u>50-1177</u> in the amount of \$860.00 to cover the above fees. A triplicate copy of this sheet is enclosed. Order No. <u>01-0041</u> . c. <input checked="" type="checkbox"/> The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. <u>50-1177</u> . A triplicate copy of this sheet is enclosed. NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137 (a) or (b)) must be filed and granted to restore the application to pending status.		
SEND ALL CORRESPONDENCE TO: Cognis Corporation, Law Dept. 2500 Renaissance Blvd., Suite 200 Gulph Mills, PA 19406		
		SIGNATURE: <u>John E. Dorsch</u> John E. Dorsch NAME ATTORNEY FOR APPLICANT 32,891 REGISTRATION NUMBER

09/744001

JC07 Rec'd PCT/PTO 18 JAN 2001

"Express Mail" mailing label number EL541613488US.

PATENT
Docket No. H 3329 PCT/US

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

RE: PCT/EP99/04836
International Filing Date: July 9, 1999
Priority Date Claimed: July 18, 1998
Applicant: Behler, et al.
Title: METHOD FOR PREPARING ALCOXYLATED CARBOXYLIC
ACID ESTERS
Applicants' Reference: H 3329 PCT/US

PRELIMINARY AMENDMENT

Commissioner for Patents
Box PCT
Washington, DC 20231

ATTN: DO/EO/US

Sir:

Before examination, in the national stage for the United States, of the above-captioned application under the Patent Convention Treaty, please amend as follows the translation supplied herewith of the application:

In the Specification:

On page 1, please delete everything above the title.

On page 1, immediately above the title please insert --TITLE OF THE INVENTION:--

On page 1, between the title and line 1, please insert --BACKGROUND OF THE INVENTION:--

On page 2, between lines 28 and 29, please insert --SUMMARY OF THE INVENTION:--

On page 3, between lines 11 and 12, please insert --DESCRIPTION OF THE INVENTION:--

On page 9, at the beginning thereof please delete "CLAIMS",

On page 9, before claim 1, please add --What is claimed is:--

**Preliminary Amendment of U.S. National Stage for International Application
PCT/EP99/04836 filed July 9, 1999**

In the Claims:

Please cancel claims 1-10.

Please add the following new claims 11-24.

11. A process for the production of alkoxylated carboxylic acid esters comprising reacting a carboxylic acid ester with an alkylene oxide having from 2 to 4 carbon atoms in the presence of a catalyst wherein the catalyst is a mixture comprised of a sodium and a potassium salt selected from the group consisting of hydroxides, oxides, carbonates, alcoholates and carboxylates wherein the weight ratio of the sodium to the potassium salt is from about 20:1 to about 1:20.

12. The process of claim 11 wherein the weight ratio of the sodium to the potassium salt is from about 10:1 to about 1:10.

13. The process of claim 12 wherein the weight ratio of the sodium to the potassium salt is from about 1:1 to about 1:5.

14. The process of claim 11 wherein the catalyst is comprised of a sodium alcoholate and a potassium salt selected from the group consisting of potassium hydroxide, potassium alcoholate, potassium carboxylate and combinations thereof.

15. The process of claim 14 wherein the sodium alcoholate is sodium methoxide.

16. The process of claim 11 wherein the catalyst is comprised of sodium hydroxide and a potassium salt selected from the group consisting of potassium hydroxide, a potassium alcoholate, a potassium carboxylate and combinations thereof.

17. The process of claim 11 wherein the catalyst is comprised of sodium hydroxide and a potassium salt selected from the group consisting of potassium hydroxide, potassium methylate, potassium butylate, potassium acetate and

**Preliminary Amendment of U.S. National Stage for International Application
PCT/EP99/04836 filed July 9, 1999**

combinations thereof.

18. The process of claim 11 wherein the amount of the catalyst is from about 0.2 to about 5% by weight based on the alkoxyated carboxylic acid ester.

19. The process of claim 11 wherein the carboxylic acid ester is an ester of a C₆₋₂₂ carboxylic acid and C₁₋₂₂ monoalcohol or a polyol having from 2 to 6 hydroxyl groups and 2 to 32 carbon atoms.

20. The process of claim 19 wherein the carboxylic acid ester is a methyl ester of C₆₋₂₂ carboxylic acid.

21. The process of claim 11 wherein the alkylene oxide is ethylene oxide.

22. The process of claim 11 wherein mole ratio of the carboxylic acid ester to the alkylene oxide is from about 1:1 to about 1:40.

23. The process of claim 11 wherein the process is carried out under autogenous pressure and at a temperature of from about 100 to about 180°C.

24. The process of claim 23 wherein the temperature is from about 160 to about 180°C.

REMARKS

Claims 11-24 are currently pending in the instant application.

The Specification has been amended to include the preferred section headings pursuant to 37 C.F.R. § 1.77. It is submitted that the amendments to the Specification made herein introduce no new matter. Their entry is therefore proper and respectfully requested. An Abstract of the Disclosure in accordance with the abstract of the corresponding international publication has been added on a separate sheet following the claims.

**Preliminary Amendment of U.S. National Stage for International Application
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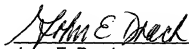
Original claims 1-10 have been canceled and replaced with new claims 11-24 solely for the purpose of improving clarity and grammar, which may suffer in translation, and not for any reason related to the statutory requirements for a patent.

New claims 11-24 have not been added in response to any rejection, nor in anticipation of any rejection related to the statutory requirements for a patent.

Applicants respectfully submit that the scope of new claims 11-24 corresponds to the scope of original claims 1-10 and that new claims 11-24 are no narrower than original claims 1-10. Furthermore, although a moot point in view of their cancellation, Applicants respectfully submit that original claims 1-10 satisfied the requirements of 35 U.S.C. § 112, as filed. New claims 11-24 are supported by the specification and no new matter has been introduced. Entry is therefore proper and respectfully requested.

Prompt examination of the instant application in view of the amendments made herein is respectfully requested.

Respectfully submitted,



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Method for Preparing Alcoxylated Carboxylic Acid Esters

This invention relates to a process for the production of alcoxylated carboxylic acid esters in the presence of a basic homogeneous catalyst mixture containing sodium and potassium compounds in a selected mixing ratio.

- 5 Alcoxylated carboxylic acid esters, preferably ethoxylated carboxylic acid methyl esters (which are also known as methyl ester ethoxylates), are known nonionic surfactants which have recently acquired considerable interest by virtue of their excellent washing performance.

- 10 They are normally produced by reacting carboxylic acid esters with alkylene oxides in the presence of basic catalysts, the alkylene oxides being inserted into the carbonyl ester compound. The insertion of alkylene oxides into the carbonyl ester compound is far more difficult than the addition of alkylene oxides onto compounds containing acidic hydrogen atoms and, accordingly, can only be achieved using special catalysts.

- 15 The use of calcined or fatty-acid-modified hydrotalcites for the ethoxylation of fatty acid esters is known from **EP-B1-0 339 425** and **EP-B1-0 523 089**. According to **DE-A1 44 46 064**, the ethoxylation of methyl esters is carried out in the presence of mixed metal oxides which have been surface-modified with metal hydroxides or metal alkoxides.
- 20 Unfortunately, these processes have a number of disadvantages. The use of heterogeneous catalysts, i.e. catalysts insoluble in the reaction mixture, involves problems of a technical nature because, unlike a liquid, the solid cannot be introduced into the reactor via an automatic metering system, but normally has to be scooped into the reactor by hand. Separation of the
- 25 catalyst is also problematical because the catalyst generally consists of such fine particles that special filter candles have to be used for filtration. However, the catalyst cannot be left in the end reaction product either

because otherwise clouding and sedimentation can occur.

EP-A2-0 335 295 describes a process for the production of alkoxyolated carboxylic acid esters using a homogeneous catalyst which does not have any of the above-described disadvantages of heterogeneous catalysis. The catalysts used are alkali metal or alkaline earth metal compounds from the group of hydroxides, oxides and alcoholates which are always used individually and not in admixture. Unfortunately, catalysts such as these do not accelerate the reaction to the required extent on their own so that long reaction times are unavoidable.

According to **DE-C-196 11 508** and **EP-A1-0 783 012**, alkylene glycols and silicon compounds, respectively, are added as co-catalysts to the alkali metal and alkaline earth metal compounds. In the case of the silicon compounds, the same disadvantages that attend heterogeneous catalysis are encountered on account of the insolubility of the co-catalyst in the reaction mixture. Although, in the variant with the alkylene glycols, catalysis is homogeneous, polyethylene glycols undesirable in view of their tendency to sediment are formed because the alkylene glycols also react with the alkylene oxides used during the process.

Accordingly, the problem addressed by the invention was to provide a process for the production of alkoxyolated carboxylic acid esters which would not be attended by any of the disadvantages mentioned above and which would ensure that the catalysts used on the one hand would dissolve in the reaction product and, on the other hand, would have such high reactivity that shorter reaction times would be possible for the same product quality.

The problem stated above has been solved by the use of a basic catalyst mixture of sodium and potassium compounds in a selected mixing ratio.

In one embodiment, the present invention relates to a process for the production of alkoxyolated carboxylic acid esters by reacting C_{2-4}

alkylene oxides in the presence of a basic catalyst, characterized in that a mixture of sodium and potassium compounds from the group of hydroxides, oxides, carbonates, alcoholates and carboxylates in a ratio by weight of sodium to potassium compounds of 20:1 to 1:20 is used as the basic catalyst.

Whereas the sodium or potassium compounds on their own have only minimal activity as catalysts so that, technically speaking, long reaction times have to be accepted for the alkoxylation of the carboxylic acid esters, it has surprisingly been found that mixtures of the described sodium and potassium compounds in certain ratios have a significantly higher activity so that the same reaction takes place far more quickly.

Carboxylic acid esters

The carboxylic acid esters suitable as starting materials for the alkoxylation are esters of carboxylic acids with monoalcohols or esters of carboxylic acids with polyols. Preferred carboxylic acids are C₆₋₂₂ carboxylic acids of natural or synthetic origin, more particularly linear, saturated and/or unsaturated fatty acids, including the technical mixtures thereof which can be obtained by lipolysis from animal and/or vegetable fats and oils, for example from coconut oil, palm kernel oil, palm oil, soybean oil, sunflower oil, rapeseed oil, cottonseed oil, fish oil, bovine tallow and lard. Examples of such preferred carboxylic acids are caproic acid, caprylic acid, 2-ethylhexanoic acid, capric acid, lauric acid, isotridecanoic acid, myristic acid, palmitic acid, palmitoleic acid, stearic acid, isostearic acid, oleic acid, elaidic acid, petroselinic acid, linoleic acid, linolenic acid, elaeostearic acid, arachic acid, gadoleic acid, behenic acid and/or erucic acid.

Suitable monoalcohols are primary alcohols containing 1 to 22 carbon atoms which may be saturated and/or unsaturated. Examples of suitable monoalcohols are methanol, ethanol, propanol, butanol, pentanol

and the hydrogenation products of the above mentioned C_{6-22} carboxylic acids. The methyl esters of the C_{6-22} carboxylic acids are particularly preferred.

- Suitable polyols contain 2 to 6 hydroxyl groups in the molecule and 2 to 32 carbon atoms. Examples of suitable polyols are ethylene glycol, 1,2-propylene glycol, 1,2-butylene glycol, neopentyl glycol, glycerol, diglycerol, triglycerol, trimethylol propane, pentaerythritol and sorbitol. If carboxylic acid esters of polyols are used, they may be present as full esters or as partial esters or as technical ester mixtures containing partial esters, more particularly in the form of glycerides.

In a particularly preferred embodiment of the invention, methyl esters of C_{6-22} carboxylic acids, more particularly methyl esters of C_{12-18} carboxylic acids, are used as starting compound.

15 Alkylene oxides

The alkylene oxides used are ethylene oxide, propylene oxide and/or butylene oxide, preferably ethylene oxide and/or propylene oxide, more particularly ethylene oxide on its own.

20 Catalysts

- The basic catalyst used is a mixture of sodium compounds and potassium compounds from the group of hydroxides, oxides, carbonates, alcoholates and carboxylates. Suitable alcoholates are the corresponding sodium and potassium compounds derived from monoalcohols containing 1 to 6 carbon atoms and preferably up to 4 carbon atoms, for example from methanol, ethanol, propanol, n-butanol or t-butanol. Suitable carboxylates are the corresponding sodium and potassium compounds derived from monobasic carboxylic acids containing 1 to 22 carbon atoms and preferably 2 to 4 carbon atoms, for example from acetic acid, propionic acid, butyric acid and from the C_{6-22} carboxylic acids already described in connection

with the carboxylic acid esters.

The sodium and potassium compounds in the catalyst mixtures may come from the same group, i.e. both are, for example, hydroxides or alcoholates, or from different groups, i.e. one compound is an alcoholate and the other a carboxylate or hydroxide. In one embodiment of the invention, the catalyst mixture contains a sodium alcoholate, more particularly sodium methylate. Accordingly, catalyst mixtures consisting of sodium alcoholates, more particularly sodium methylate, and potassium hydroxide, potassium alcoholates and/or potassium carboxylates, more particularly potassium hydroxide, potassium methylate, potassium butylate (from t-butanol) and/or potassium acetate, are preferred. In another embodiment of the invention, the catalyst mixture consists of sodium hydroxide and potassium hydroxide, potassium alcoholates and/or potassium carboxylates, more particularly potassium hydroxide, potassium methylate, potassium butylate (from t-butanol) and/or potassium acetate.

The mixing ratio of sodium to potassium compounds in the catalyst mixture to be used in accordance with the invention is 20:1 to 1:20, preferably 10:1 to 1:10 and more preferably 1:1 to 1:5. In the context of the present invention, the mixing ratio is defined as the ratio by weight of the sodium compound to the potassium compound.

The catalyst mixture otherwise contains no other catalytically active constituents.

The catalyst mixture is preferably used in quantities of 0.2 to 5% by weight, based on alkoxyated carboxylic acid ester as reaction product. Although larger quantities by weight may be used, they are uneconomical. Smaller quantities by weight may also be used but lengthen the reaction time.

Alkoxylation

The alkoxylation reaction may be carried out in known manner. To

this end, the carboxylic acid ester is normally introduced into a stirrer-equipped autoclave and the homogeneous catalyst is subsequently added, for example as a solid or in the form of a solution, preferably in water and/or in methanol. It has proved to be of advantage to purge the autoclave thoroughly with nitrogen before the reaction to remove all traces of atmospheric oxygen and to remove methanol used as solvent by evacuation. The autoclave is then heated. The alkoxylation reaction is carried out at temperatures of preferably 100 to 180°C and more preferably 160 to 180°C. The alkylene oxide, which may be ethylene oxide, propylene oxide or mixtures of both, is introduced into the reactor by a siphon. The autogenous pressure can rise to about 5 bar. The alkylene oxide, preferably ethylene oxide, is preferably used in a quantity of on average 1 to 40 moles and more preferably 5 to 15 moles per mole of carboxylic acid ester. In other words, the molar ratio of carboxylic acid ester to alkylene oxides is in the range from 1:1 to 1:40 and preferably in the range from 1:5 to 1:15. The addition of the alkylene oxide is statistical, i.e. the insertion is not a highly selective reaction in which 1 mole of fatty acid alkyl ester reacts with exactly n moles of alkylene oxide. Instead, a complex mixture of esters alkoxylation to different degrees is obtained. The reaction is over when the pressure in the reactor falls to about 0.5 bar. For safety reasons, it is advisable to stir the mixture for another 30 minutes before the reactor is cooled and vented. If desired, the alkaline catalyst can be neutralized by the addition of acids, for example phosphoric acid, acetic acid, lactic acid or the like.

25

Commercial Applications

The alkoxylation carboxylic acid esters can be produced in a relatively short reaction time of under 4 hours by the process according to the invention. Compared with the use of sodium methylate, for example, on its own as the basic catalyst, this means that the reaction time is halved,

30

more particularly for comparable product quality.

The present invention relates to the use of the alkoxylated carboxylic acid esters produced by the process according to the invention as a surfactant for the production of laundry detergents, dishwashing detergents

5 and cleaners, more particularly manual dishwashing detergents, multipurpose cleaners, floor cleaners, glass cleaners, dishwasher detergents and liquid laundry detergents.

The alkoxylated carboxylic acid esters are nonionic surfactants with high cleaning performance and, accordingly, may be used in combination

10 with other anionic, nonionic and/or cationic surfactants in laundry detergents, dishwashing detergents and cleaners, preferably in quantities of 0.5 to 30% by weight, based on laundry detergent, dishwashing detergent or cleaner. The corresponding detergents/cleaners may additionally contain typical ingredients in typical quantities.

Examples

General procedure. 290 g (1.34 mole) of lauric acid methyl ester were introduced into a 1-liter stirred autoclave and the proposed quantity of

20 catalyst mixture was added. The autoclave was closed and was then alternately purged with nitrogen and evacuated for 30 mins. at 100°C three times to rule out the presence of atmospheric oxygen. The reaction mixture was then heated to 165°C-175°C under a nitrogen blanket and 710 g (16.13 moles) of ethylene oxide were introduced in portions, the

25 autogenous pressure initially rising to 3.5 bar. The reaction was continued until the pressure had fallen to 0.5 bar. After stirring for another 30 minutes, the autoclave was cooled and vented. The results of the tests are set out in Table 1. The quantities of catalysts used (in % by weight) are based on the end product (ethoxylated lauric acid ester).

Table 1. Catalyst composition
Ethoxylation of lauric acid methyl ester

Ex.	Sodium compound	% by weight	Potassium compound	% by weight	Reaction time (h)
1.	Sodium methylate ¹	0.5	Potassium acetate	0.5	3.5
2	Sodium methylate ¹	0.5	Potassium acetate	3.0	2.0
3	Sodium methylate ¹	0.5	Potassium methylate ¹	0.5	2.25
4	Sodium methylate ¹	0.5	Potassium-t-butyrate	0.5	2.25
5	Sodium hydroxide ²	0.5	Potassium hydroxide ²	0.5	3.5
C1	Sodium methylate ¹	2.0	None	-	8.5
C2	Sodium methylate ¹	0.5	Sodium acetate	0.5	Over 8.5; terminated ³
C3	Potassium hydroxide ²	0.5	Potassium acetate	3.0	Over 8.5; terminated ³

¹ 30% by weight solution in methanol

² 50% by weight solution in water

³ the reaction was terminated if the uptake of ethylene oxide was not complete (no drop in pressure to 0.5 bar)

CLAIMS

1. A process for the production of alkoxyated carboxylic acid esters by reacting alkylene oxides containing 2 to 4 carbon atoms in the presence of a basic catalyst, characterized in that a mixture of sodium and potassium compounds from the group of hydroxides, oxides, carbonates, alcoholates and carboxylates in a ratio by weight of sodium to potassium compounds of 20:1 to 1:20 is used as the basic catalyst.
2. A process as claimed in claim 1, characterized in that the sodium and potassium compounds are used in a mixing ratio of 10:1 to 1:10 and preferably 1:1 to 1:5.
3. A process as claimed in claim 1 or 2, characterized in that sodium alcoholates, preferably sodium methylate, are used in admixture with potassium hydroxide, potassium alcoholates and/or potassium carboxylates.
4. A process as claimed in any of claims 1 to 3, characterized in that sodium hydroxide is used in admixture with potassium hydroxide, potassium alcoholates and/or potassium carboxylates, preferably potassium hydroxide, potassium methylate, potassium butylate and/or potassium acetate.
5. A process as claimed in any of claims 1 to 4, characterized in that the catalyst mixture is used in quantities of 0.2 to 5% by weight, based on alkoxyated carboxylic acid ester.
6. A process as claimed in any of claims 1 to 5, characterized in that esters of C₆₋₂₂ carboxylic acids and C₁₋₂₂ monoalcohols or with polyols containing 2 to 6 hydroxyl groups and 2 to 32 carbon atoms, more particularly methyl esters of C₆₋₂₂ carboxylic acids, are used as the carboxylic acid esters.
7. A process as claimed in any of claims 1 to 6, characterized in that ethylene oxide is used as the alkylene oxide.
8. A process as claimed in any of claims 1 to 7, characterized in that

the carboxylic acid esters and alkylene oxides are used in a molar ratio of 1:1 to 1:40.

9. A process as claimed in any of claims 1 to 8, characterized in that the reaction is carried out under autogenous pressure at temperatures in the range from 100 to 180°C and preferably at temperatures in the range from 160 to 180°C.
- 5 10. The use of the alkoxyated carboxylic acid esters obtained by the process claimed in claims 1 to 9 as a surfactant for the production of laundry detergents, dishwashing detergents and cleaners.

ABSTRACT OF THE DISCLOSURE

Alkoxyalted carboxylic acid esters are made by a process which comprises reacting a carboxylic acid ester with an alkylene oxide having from 2 to 4 carbon atoms in the presence of a catalyst wherein the catalyst is a mixture comprised of a sodium and a potassium salt selected from the group consisting of hydroxides, oxides, carbonates, alcoholates and carboxylates wherein the weight ratio of the sodium to the potassium salt is from about 20:1 to about 1:20.

Type a plus sign (+) inside this box ☐

0010/PTO
Rev. 6/95

U.S. Department of Commerce
Patent and Trademark Office

DECLARATION FOR UTILITY OR DESIGN PATENT APPLICATION

☐ Declaration Submitted with Initial Filing OR ☒ Declaration Submitted after Initial Filing

Attorney Docket Number

H 3329 PCT/US

First Named Inventor

BEHLER, Ansgar

COMPLETE IF KNOWN

Application Number

09/744,001

Filing Date

03/16/2001

Group Art Unit

Examiner Name

As a below named inventor, I hereby declare that:

My residence, post office address, and citizenship are as stated below next to my name.

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

METHOD FOR PREPARING ALCOXYLATED CARBOXYLIC ACID ESTERS

(Title of the invention)

the specification of which

☐ is attached hereto

OR

☒ was filed on (MM/DD/YYYY) 07/09/1999 as United States Application Number or PCT International

Application Number PCT/EP99/04836 and was amended on (MM/DD/YYYY) (if applicable).

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment specifically referred to above.

I acknowledge the duty to disclose information which is material to patentability as defined in Title 37 Code of Federal Regulations, § 1.56.

I hereby claim foreign priority benefits under Title 35, United States Code § 119(a)-(d) or § 385(b) of any foreign application(s) for patent or inventor's certificate, or § 385(a) of any PCT International application which designated at least one country other than the United States of America, listed below and have also identified below, by checking the box, any foreign application for patent or inventor's certificate, or of any PCT International application having a filing date before that of the application on which priority is claimed.

Prior Foreign Application Number(s)	Country	Foreign Filing Date (MM/DD/YYYY)	Priority Not Claimed	Certified Copy Attached? YES NO
198 32 427.8	DE	07/18/1998	<input type="checkbox"/>	<input type="checkbox"/> <input checked="" type="checkbox"/>
			<input type="checkbox"/>	<input type="checkbox"/> <input type="checkbox"/>
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			<input type="checkbox"/>	<input type="checkbox"/> <input type="checkbox"/>
			<input type="checkbox"/>	<input type="checkbox"/> <input type="checkbox"/>

☐ Additional foreign application numbers are listed on a supplemental priority sheet attached hereto:

I hereby claim the benefit under Title 35, United States Code § 119(e) of any United States provisional application(s) listed below.

Application Number(s)	Filing Date (MM/DD/YYYY)	Additional provisional application numbers are listed on a supplemental priority sheet attached hereto.
		<input type="checkbox"/>

Burden Hour Statement: This form is estimated to take 4 hours to complete. Time will vary depending upon the needs of the individual case. Any comments on the amount of time you are required to complete this form should be sent to the Chief Information Officer, Patent and Trademark Office, Washington, DC 20231. DO NOT SEND FEES OR COMPLETED FORMS TO THIS ADDRESS. SEND TO: Assistant Commissioner for Patents, Washington, DC 20231.

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DECLARATION

Page 2

I hereby claim the benefit under Title 35, United States Code §120 of any United States application(s), or §365(c) of any PCT international application designating the United States of America, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT international application in the manner provided by the first paragraph of Title 35, United States Code §112.1 acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations §1.56 which became available between the filing date of the prior application and the national or PCT international filing date of this application.

U.S. Parent Application Number	PCT Parent Number	Parent Filing Date (MM/DD/YYYY)	Parent Patent Number (if applicable)
	PCT/EP99/04836	07/09/1999	

☐ Additional U.S. or PCT international application numbers are listed on a supplemental priority sheet attached hereto.

As a named inventor, I hereby appoint the following attorney(s) and/or agent(s) to prosecute this application and to transact all business in the Patent and Trademark Office connected therewith:

☐ Firm Name Customer Number or label

☒ List Attorney(s) and/or agent(s) name and registration number below:

Name	Registration Number	Name	Registration Number
John E. Drach	32,891	Aaron R. Ettelman	42,516
Steven J. Trzaska	36,296	Henry E. Millson, Jr.	78,980

☐ Additional attorney(s) and/or agent(s) named on a supplemental sheet attached hereto.

Please direct all correspondence to: ☒ Customer Number **23657** or label ☐ OR ☒ Fill in correspondence address below

Name	John E. Drach		
Address			
Address			
City	State	ZIP	
Country	Telephone	Fax	
	610-278-4925	610-278-6548	

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Name of Sole or First Inventor:

☐ A petition has been filed for this unsigned

Given Name	Ansgar	Middle Initial		Family Name	Behler	Suffix e.g. Jr.	
Inventor's Signature	<i>Dans B. Behler</i>				Date	01/31/2001	
Residence: City	Bottrop	State		Country	Germany	Citizenship	Germany
Post Office Address	Siegfriedstrasse 80 DEX						
Post Office Address							
City	45240 Bottrop	State		Zip		Country	Germany
Applicant Authority							

☒ Additional inventors are being named on supplemental sheet(s) attached hereto

DECLARATION

ADDITIONAL INVENTOR(S):
Supplemental Sheet

Name of Additional Joint Inventor, if any:

☐ A petition has been filed for this unsigned inventor

Given Name	Almud	Middle Initial		Family Name	Folge	Suffix e.g. Jr.	
Inventor's Signature	Almud Folge					Date	01/13/2011
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Post Office Address	Locher Weg 37						
Post Office Address	DEx						
City	40764 Langenfeld	State		Zip		Country	Germany
						Applicant Authority	

Name of Additional Joint Inventor, if any:

☐ A petition has been filed for this unsigned inventor

Given Name		Middle Initial		Family Name		Suffix e.g. Jr.	
Inventor's Signature						Date	
Residence: City		State		Country		Citizenship	
Post Office Address							
Post Office Address							
City		State		Zip		Country	
						Applicant Authority	

Name of Additional Joint Inventor, if any:

☐ A petition has been filed for this unsigned inventor

Given Name		Middle Initial		Family Name		Suffix e.g. Jr.	
Inventor's Signature						Date	
Residence: City		State		Country		Citizenship	
Post Office Address							
Post Office Address							
City		State		Zip		Country	
						Applicant Authority	

Name of Additional Joint Inventor, if any:

☐ A petition has been filed for this unsigned inventor

Given Name		Middle Initial		Family Name		Suffix e.g. Jr.	
Inventor's Signature						Date	
Residence: City		State		Country		Citizenship	
Post Office Address							
Post Office Address							
City		State		Zip		Country	
						Applicant Authority	

☐ Additional inventors are being named on supplemental sheet(s) attached hereto